MICHIGAN DEPARTMENT OF
ENVIRONMENT, GREAT LAKES, AND ENERGY

New Control Strategy Approaches for Ozone Attainment

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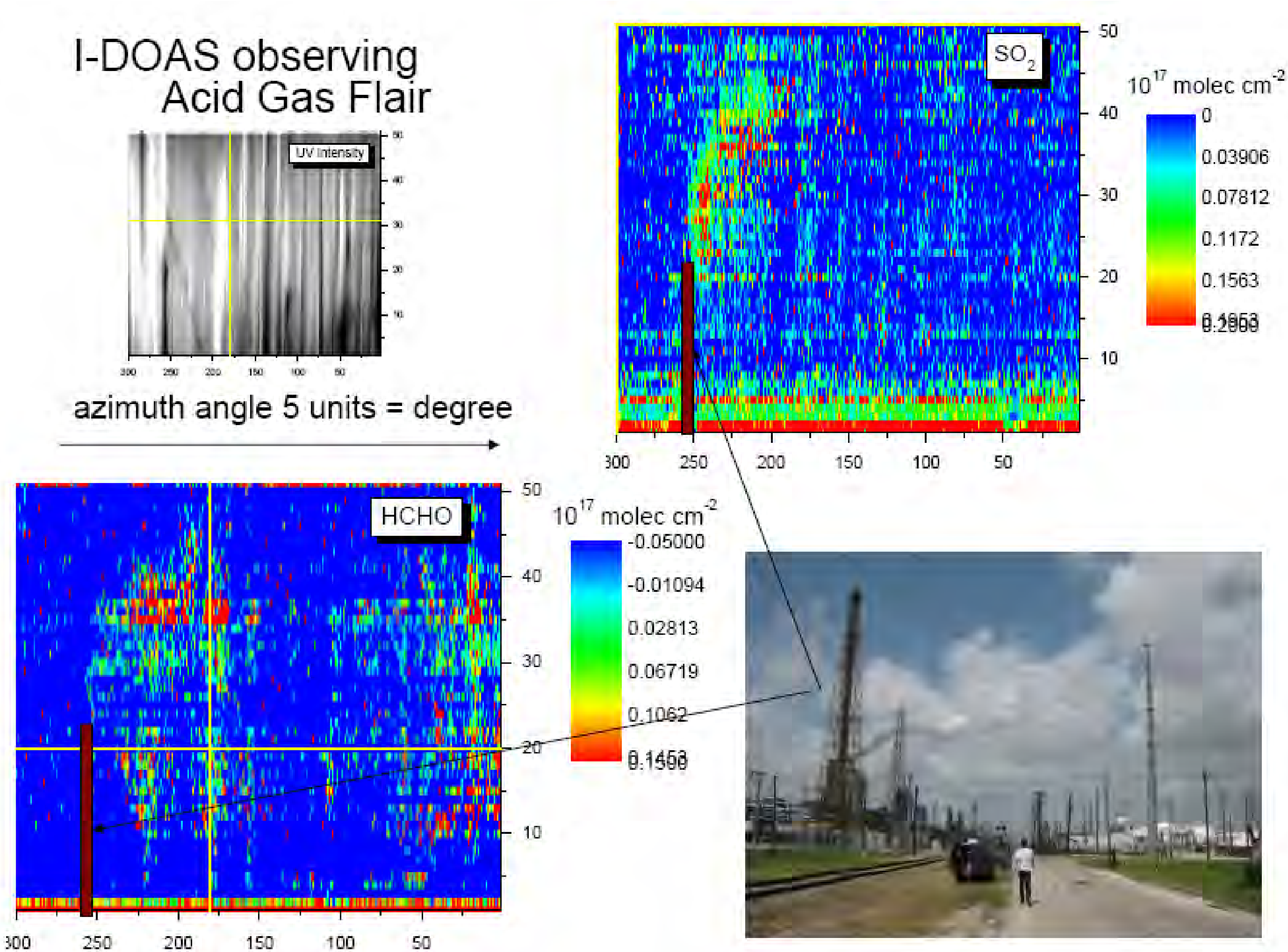
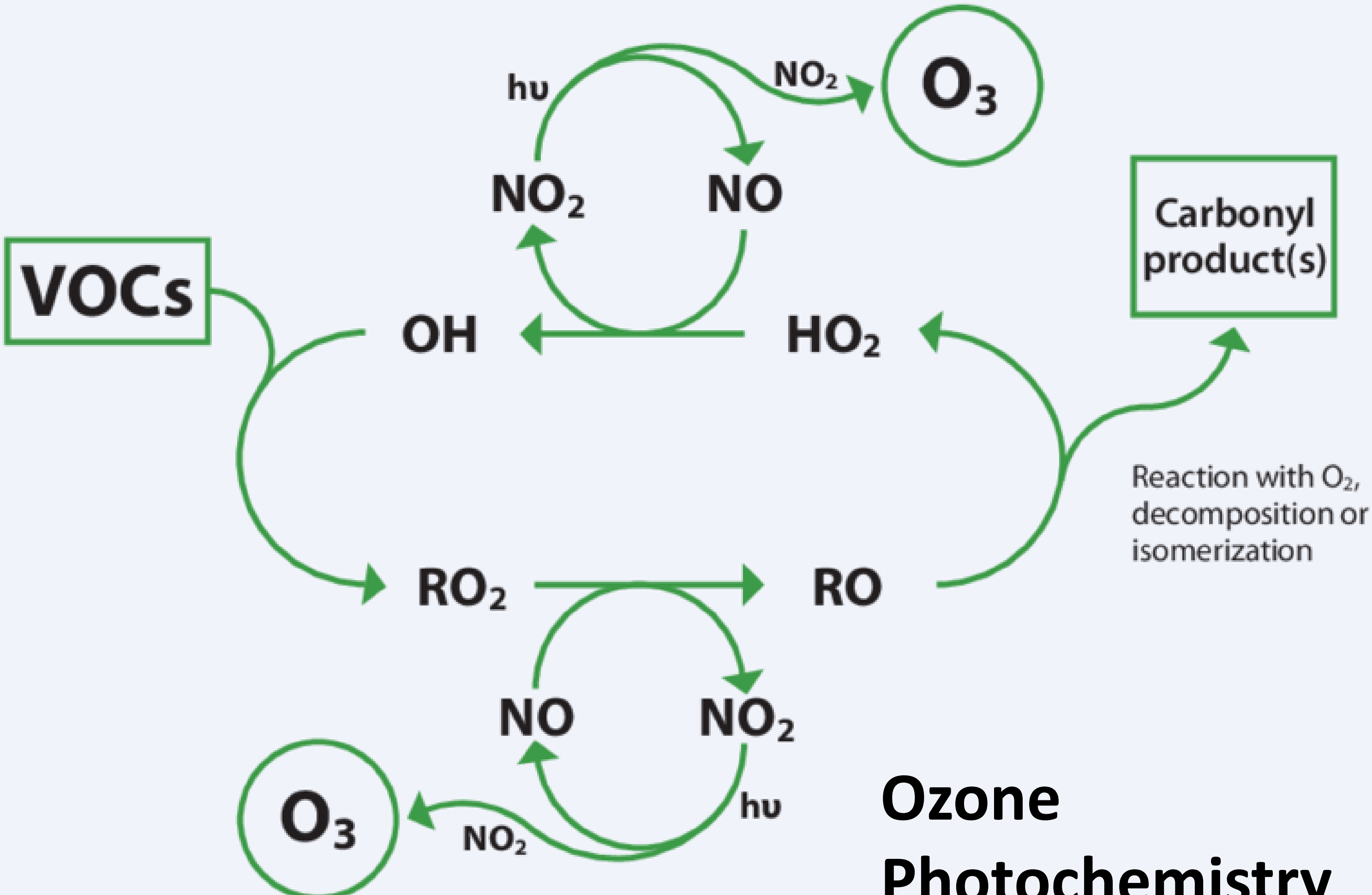
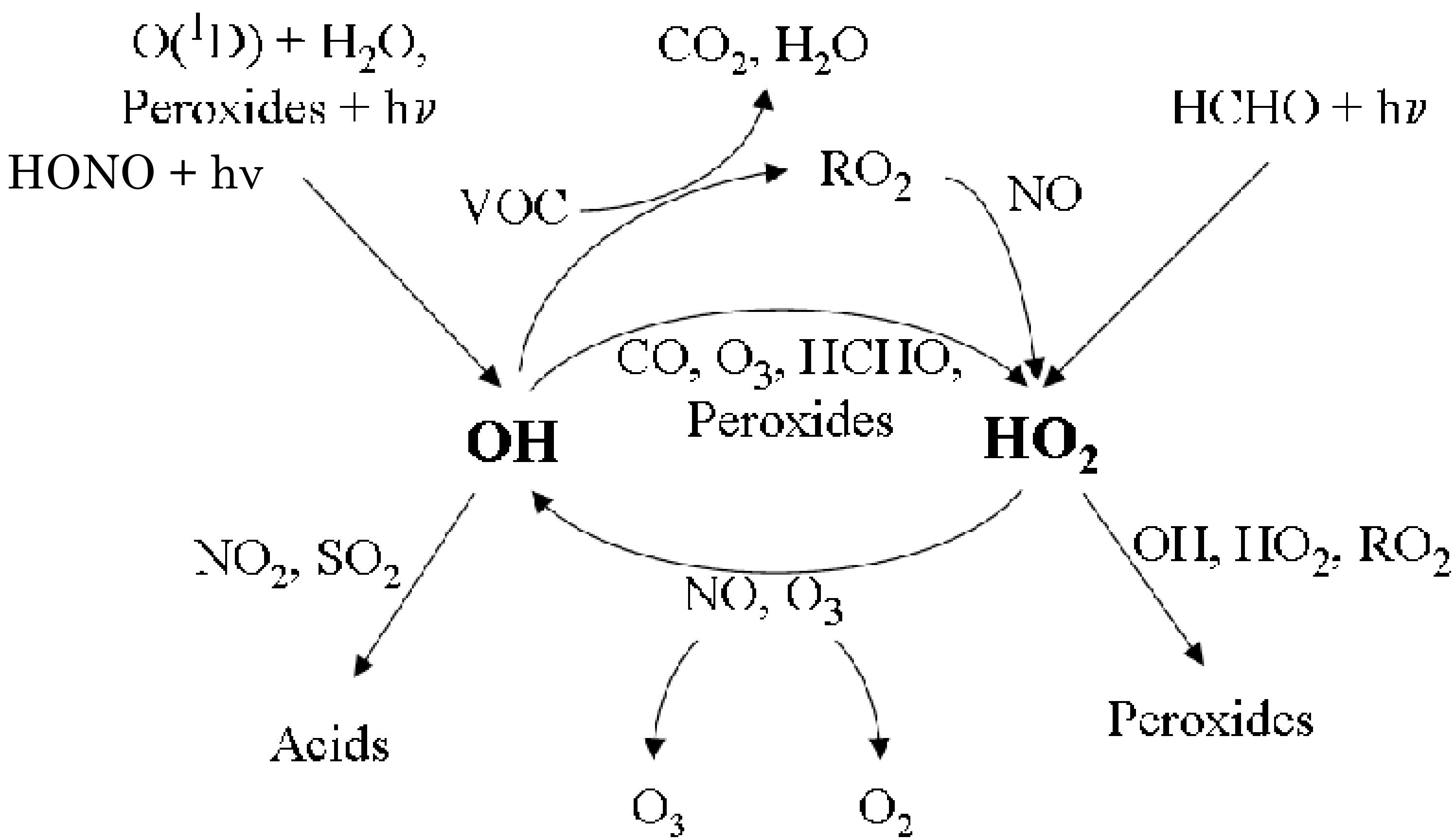
1. Primary Formaldehyde





Ozone Photochemistry

Radical Precursors



HCHO as a Radical Precursor

- In the past, radicals were extremely difficult to measure. **Laser-induced fluorescence** has enabled atmospheric radical budgets to be quantified.
- Most VOCs are sources of **internal radicals** only. Without enough **external radical sources**, ozone production is limited.
- **Radical precursors are the "match" that lights the "flame"** of ozone reaction chains by supplying external radicals.
- Besides H_2O, a main source of external radicals is **formaldehyde (HCHO).**
- HCHO has both **primary** (combustion emissions) and **secondary** (formation in the atmosphere due to VOC decomposition) sources.

Evidence for Primary Formaldehyde

- **Point source inventories often underestimate or ignore primary formaldehyde** from incomplete combustion. A Houston modeling study suggested that **uncounted primary formaldehyde could increase peak 1-hr ozone by up to ~30 ppb** (Vizuete et al., 2006).

- Aircraft and ground-based measurements during **TexAQS II** in 2006 indicated **large combustion sources of HCHO**, such as industrial flares (Olaguer et al., *J. Air & Waste Manage. Assoc.*, **59**, 1258-1277, 2009).

- Remote sensing studies during the 2009 **Study of Houston Atmospheric Radical Precursors (SHARP)** verified the existence of significant primary HCHO (Olaguer et al., *J. Geophys. Res.-Atmos.*, **119**, 2597-2610, 2014).

- Analysis of both mobile laboratory and aircraft data demonstrated that **HCHO:CO molar ratios in large combustion plumes were around 2–10%**.

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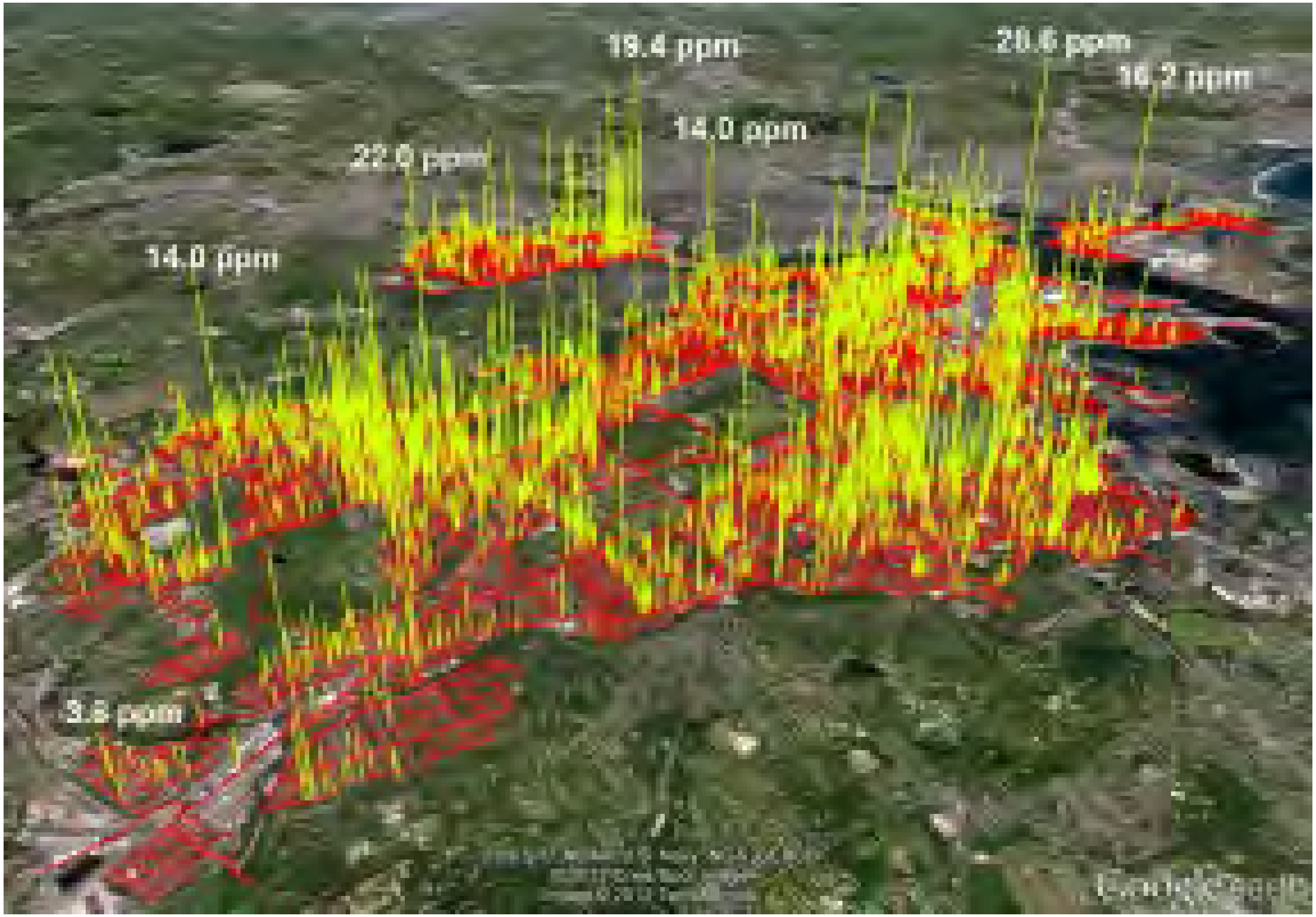
What are the Implications of Primary HCHO?

- Air quality models may underestimate ozone production due to missing primary HCHO (e.g., from NG-fired EGUs and stationary engines, landfill and refinery flares, etc.).

- The simulated effectiveness of combustion control strategies in O_3 attainment demonstration models may be enhanced by including more primary HCHO.

2. Underground Pipeline Leaks



Methane Leaks in Boston Measured by Phillips et al. (2013)

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Why Pipeline Leaks?

- Emissions from natural gas distribution and end use may be 2-3 times larger than predicted by existing inventory methodologies and industry reports (McKain et al., *PNAS, 112: 1941-1946, 2015*).

- Phillips et al. (*Environmental Pollution, 173:1-4, 2013*) identified 3356 methane leaks in Boston with concentrations exceeding up to 15 times the global background level.

- Urban areas with corrosion-prone distribution lines leak ~25-fold more methane than cities with more modern pipeline materials (Fischer et al., *Environ, Sci. Technol., 51: 4091-4099, 2017*).

Potential Leak Impacts

- Normally, methane is only considered as a global tropospheric ozone precursor and not a VOC due to its long lifetime (~9 years).
- Modeling by USEPA ~4 years ago suggested that large pipeline leaks of methane in urban areas may lead to significant increases in local ozone (Dr. Rohit Mathur, USEPA, personal communication).
- There may be wet natural gas, crude oil, or refined product in pipelines resulting in fugitive emissions of more reactive VOCs with greater ozone formation potential (e.g., aromatics).

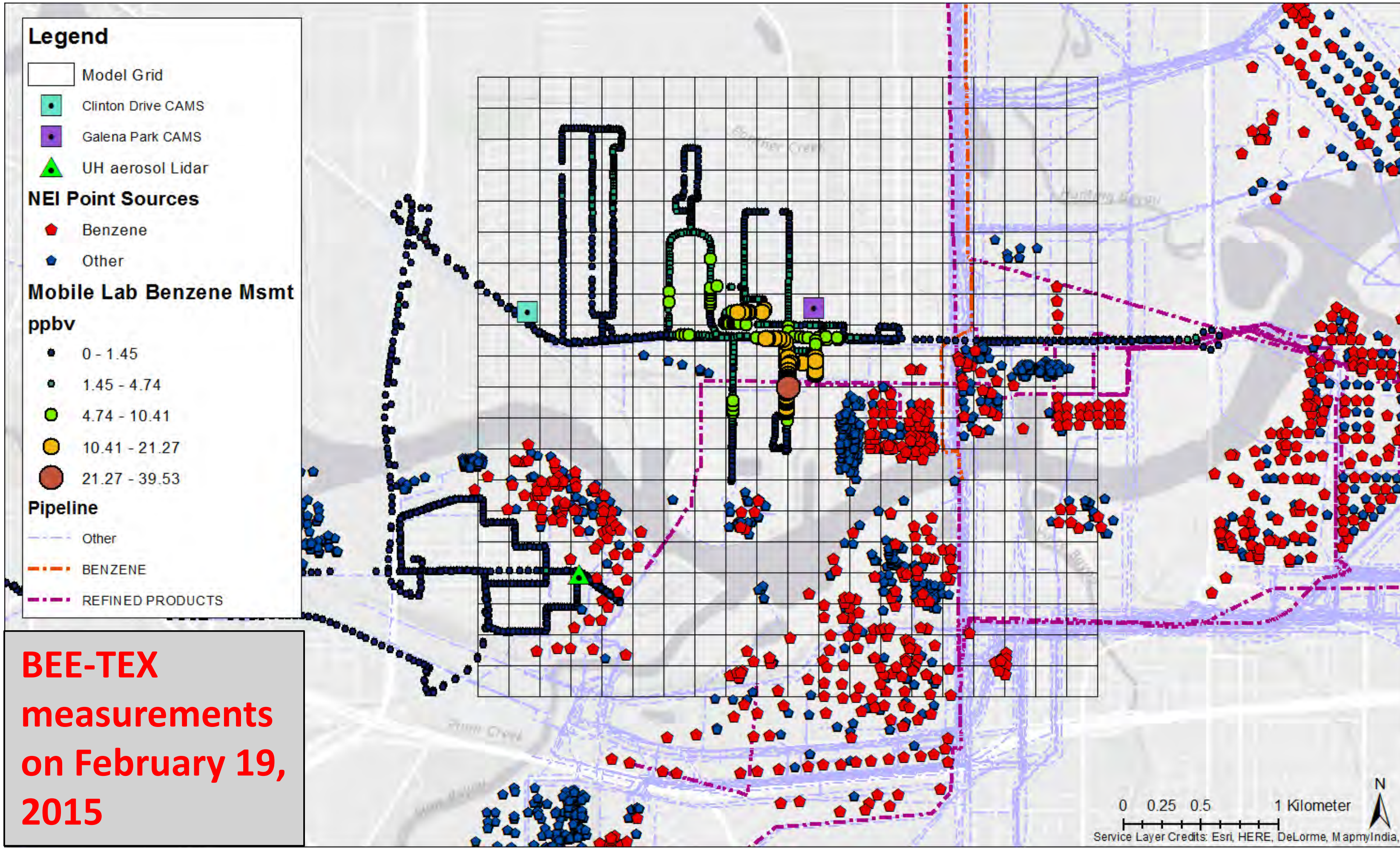
Benzene and other Toxics Exposure Study (BEE-TEX)

- **Benzene and other Toxics Exposure Study (BEE-TEX)** occurred during February 2015 in the Houston Ship Channel.

- Three mobile labs equipped with **Proton Transfer Reaction—Mass Spectrometry** (PTR-MS), plus GPS and meteorological measurements.

- **Real time source attribution and emissions quantification** (within 1 hr of measurements) based on high-resolution inverse modeling with a **3D microscale Eulerian grid transport model**.

Pipeline Network, Point Sources, and Mobile Lab Measurements of Benzene in Galena Park, Texas



BEE-TEX measurements on February 19, 2015

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Feb 19, 2015 Galena Park Benzene Total Domain Emissions (kg/hr)

Time Period	Point Sources	Pipelines	Total Emissions
Afternoon	16.43	34.73	51.16
Evening	5.59	10.69	16.29
2011 NEI	8.27	0	8.27

Olaguer, et al. (2016), *J. Air Waste Manage. Assoc.*, *66*, 164–172.

What Could Be Done About Pipelines?

- Better Leak Detection and Repair (LDAR) technology and practices.
- Incorporation of fugitive emissions from pipelines into inventories.
- High resolution modeling of ozone impacts of pipeline leaks and relevant control strategies.

3. Intermodal Transportation

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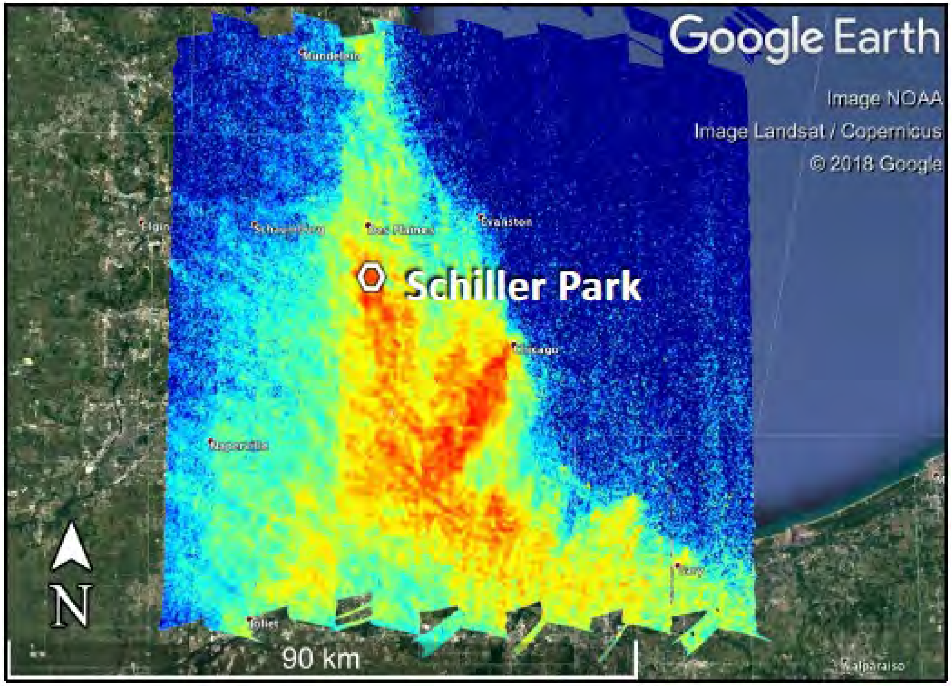
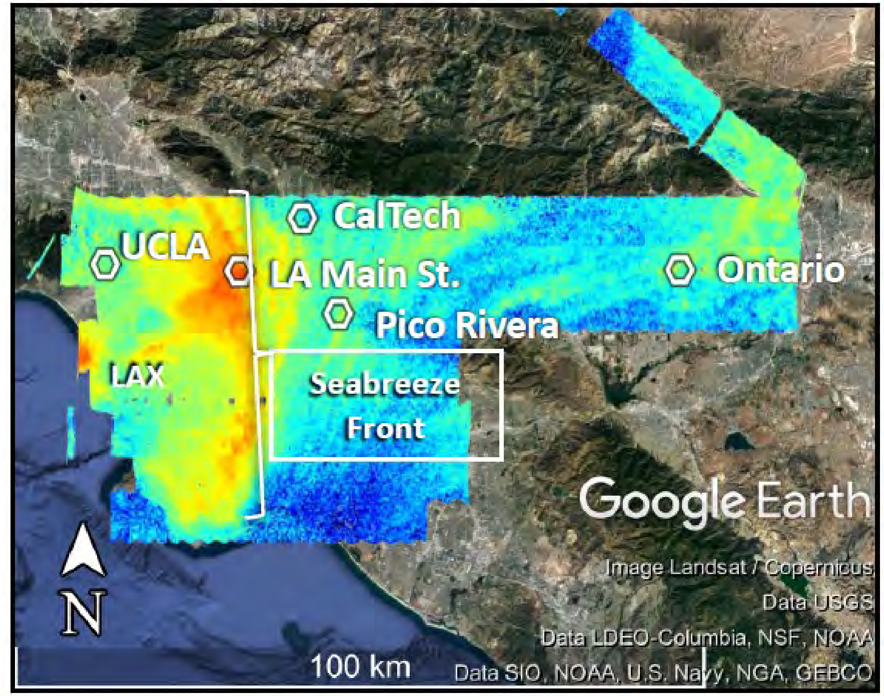
Motivations

- After thirty years of air quality improvements, much of the "low-hanging" fruit of widespread, relatively low-cost air quality control strategies has been exhausted.
- Improved vehicle fuel efficiencies and catalytic converters have significantly reduced NOx and VOC emissions.
- Transportation still generates 61% of U.S. NOx emissions.
- Heavy Duty Diesel Vehicles (HDDVs) are responsible for 24% of transportation NOx emissions, 15% of all NOx emissions.
- U.S. domestic freight tonnage may double to 30 billion tons by 2050 (AASHTO, 2010). This will expand vehicle activity at urban ports, as well as on roadways.

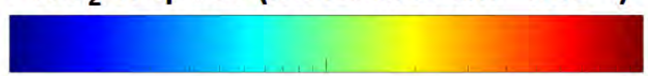
20170601 Morning Chicago Raster



Nominal: 250 x 250 m



NO_2 TropVCs (molecules cm^{-2} $x10^{15}$)



NASA deployed an airborne UV/Visible spectrometer, GeoTASO, in May-June 2017 to yield high resolution NO_2 Tropospheric Vertical Column (TropVC) data (Judd et al., 2019).

Note intense NOx plumes at O'Hare (near Schiller Park) and LAX airports!

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Potential Targets in SE Michigan

- Port of Detroit, international bridge HD truck traffic emissions (e.g., certification program)
- Marine vessel emissions (e.g., port speed limits)
- Railroad emissions (e.g., grants for engine retrofits/replacement)
- DTW airport and Selfridge airbase emissions (e.g., General Conformity)

Modeling and Measurements

- Five-person Steering Committee to address data needs for: 1) 179B Petition, and 2) O_3 attainment demonstration
- Still awaiting official USEPA guidance on how to structure a 179B Petition
- Initial discussion will focus on:
  - Ensuring accurate meteorological simulations for Southeast Michigan, Windsor, and Sarnia
  - Source apportionment data for emissions and ozone model evaluation, and possible 179B petition focused on Canadian source contributions

Summary and Conclusion

- Low-hanging fruit is exhausted, so we need entirely new control strategy approaches.
- New measurement technologies have revealed interesting un(der)controlled emission sources.
- In-depth understanding and advanced representation of ozone model physics and chemistry may help to design new control strategies.

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